Letters to the Editor

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The Exchange Integral in the 3*d* Shell

HARVEY KAPLAN
*Department of Physics, University of California, Berkeley, California**
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DURING the past twenty years there has been considerable speculation as to the sign of the Heitler-London exchange integral,

$$J=2\int \psi_m^*(1)\psi_n^*(2)\left[\frac{1}{r_{mn}}+\frac{1}{r_{12}}-\frac{1}{r_{1m}}-\frac{1}{r_{1n}}\right]\psi_n(1)\psi_m(2)d\tau_1 d\tau_2$$

for the known ferromagnetic elements. The question arises in connection with Heisenberg's theory of ferromagnetism,[1] which requires this integral to have a positive sign. In 1930, Slater[2] argued that two atoms having large internuclear separation and unfilled inner shells (both of which requirements are fulfilled for the ferromagnetics) should have a positive J. The argument was extended somewhat further by Bethe,[3] who stressed the importance of the angular dependence of the electronic wave functions. Since that time doubt as to the positive sign of J has been expressed by Slater[4] and more recently by Zener.[5]

Not much has been done in the way of actually computing exchange integrals. Bartlett[6] showed that 2 atoms each having one $2p$ electron would be most strongly bound in the triplet state, each electron having a space function with a node on the line joining the atoms,

$$\psi(r,\theta)=cr\exp(-r/2r_0)\cos\theta.$$

The positive sign he obtained for the exchange integral was an aid in explaining the paramagnetism of the oxygen molecule.

Wohlfarth[7] calculated J for iron, using spherically symmetric wave functions. In view of the predominant importance of angular dependence,[3,6] the negative sign he obtained for J is of little significance.

The purpose of the present letter is to report the result of a calculation of the exchange integral for two atoms each having one $3d$ electron. The wave function used was of the form

$$\psi(r,\theta)=cr^2\exp(-r/2r_0)P_2(\cos\theta).$$

Bartlett's[6] method of calculation was used throughout. In order to find the total energy of the state the Coulomb integral

$$C=2\int |\psi_m(1)|^2|\psi_n(2)|^2\left[\frac{1}{r_{mn}}+\frac{1}{r_{12}}-\frac{1}{r_{1n}}-\frac{1}{r_{2m}}\right]d\tau_1 d\tau_2$$

was also calculated. The $3d$ binding energy is $E=(J-C)/S$, where $S^{\frac{1}{2}}=\int\psi_m^*(1)\psi_n(1)d\tau_1$. Both J and C appear as functions of $\alpha=R/r_0$, where R is the internuclear separation. C and J were tabulated for various values of α using Bartlett's tables of $f_1(m,\alpha)$ and $A_m(l,\alpha)$. Because of enormous subtraction effects, the accuracy of the results, particularly for J, was severely limited; for example, at $\alpha=6$, $Jr_0=0.004\pm0.002$ (atomic units) probable error. However, since the accuracy of C was not quite so poor and $|C|\gg|J|$ it was possible to determine the general variation of E with α. There is maximum binding for α between 5 and 6 of about $0.2/r_0$ atomic units. At $\alpha=10$, E has fallen to about forty percent of its maximum value. This is a less rapid decrease than that obtained by Bartlett for $2p$ electrons, which is reasonable since the $3d$ function are more sharply concentrated between the nuclei.

In order to remove any uncertainty as to the sign of J, it was recalculated after first extending Bartlett's tables a few places and making a few minor corrections. This was done only for $\alpha=6$ (maximum binding) since the work is tedious. The final value obtained is $J=0.00552/r_0$ atomic units or $0.150/r_0$ ev. If the value 0.25 is used for r_0, corresponding to a reasonable fit to the $3d$ iron function, $J=0.60$ ev. This value is a factor of ten larger than ferromagnetic exchange integrals.

Although the present calculation serves to support the Heisenberg theory of ferromagnetism, several qualifications of its significance must be mentioned. The radial function used is a poor approximation to a true $3d$ function. Furthermore, at maximum binding the reasonable one-exponential fit $r_0=0.25$ corresponds to the very small internuclear separation of $\sim$0.75 Angstroms. The rough calculation indicates that J almost certainly remains positive out to about twice this separation ($\alpha=10$), but this is still considerably short of the nearest neighbor spacing in ferromagnetics. The $4s$ electrons have been neglected. The effect of the other nearest neighbor atoms has been neglected also.

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Radioactivity of $Eu^{152,154}$

ROBERT KATZ AND MILFORD R. LEE
Kansas State College, Manhattan, Kansas
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ALTHOUGH the spectrographic analysis accompanying several enriched samarium isotopes from Oak Ridge contained $\leq$0.6 percent europium, some of the more intense internal conversion lines attributed to the disintegration of europium[1] have been found photographically with neutron-activated samarium isotopes in a 180° focusing β-ray spectrograph. Conversion lines of some gamma-rays designated by Cork *et al.*[1] as 122.0 (K, L, M), 244.3 (K, L), and 343.8 (K, L) kev were found on spectrograms of neutron-irradiated Sm^{150}, while conversion lines of these three gamma-rays, together with the K conversion line of the 123.2-kev gamma-ray, were found associated with neutron-irradiated Sm^{154}. Our energies for these gamma-rays were 121.2, 243.8, 343.8, and 122.4 kev, respectively. The spectrograph was calibrated with Cs^{137}. Assuming that the europium impurity was enriched in nearly the same way as the samarium, the abundance ratio of Eu^{151}:Eu^{153} would be markedly different in the two samples, being 6:1 in the Sm^{150} enrichment and 1:16 in the Sm^{154} enrichment. In addition to these long lived conversion electrons, hard gamma-rays (half-life$>$1 yr) of energy 990 kev in the Sm^{150} sample and 1170 kev in the Sm^{154} sample were found by absorption, in lead, in a geometry calibrated with Cs^{137} and Co^{60}.

The recently revised gamma-ray assignments in Eu^{152} and Eu^{154} by Keller and Cork,[2] insofar as they refer to the transfer of the highest energy (1116 kev) gamma-ray to Eu^{154} and of the 344-kev gamma-ray to Eu^{152}, are in good agreement with our data. These results appear to be in disagreement with Fowler and Schreffler[3] who have reported finding coincidences between conversion electrons from the 123- and 344-kev gamma-rays. Our data indicate that the 122-, 244-, and 344-kev gamma-rays are associated with the disintegration of Eu^{152} while the 123-kev gamma-ray appears to be associated with the disintegration of Eu^{154}.